DETECTION AND INTERPRETATION OF COLLISIONAL TRANSFER AND ROTATIONAL ANISOTROPY FINGERPRINTS IN RESONANT FOUR-WAVE MIXING SPECTRA.

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Coherent responses produced by resonant four-wave mixing (RFWM) in a weakly absorbing medium carry valuable information on the intrinsic properties and dynamics of the quantum states involved. Here, two aspects of RFWM applications are highlighted. First, the Two-Color (TC) version of RFWM was found to be a unique spectroscopic tool to directly trace collisional state-to-state transfer in isotropic gaseous media, both in the frequency[a] and time[b] domains. Second, the RFWM techniques appeared to be very useful for studies of the rotational anisotropy[c]. Here we report new experimental one-color RFWM spectra of the OH radicals produced by laser photolysis of H_2O_2 at 266 nm. Polarization dependence and Doppler line structure of the spectra show clear evidence of the pronounced anisotropy of angular momentum (**j**) and velocity (**v**) distributions as well as on the **j-v** correlation. The obtained results directly point to the pronounced OH helicity (i.e. $\mathbf{j}\|\mathbf{v}$) which yet remained beyound the reach of purely optical means. For all mentioned cases, the line-shape theory[d] is an optimal tool to derive compact expressions for the RFWM signals.

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